Resonant light scattering by fractal clusters

Vladimir M. Shalaev*
Department of Chemistry, University of Toronto, Toronto, Ontario, Canada M5S 1A1

R. Botet and R. Jullien
Laboratoire de Physique des Solides, Université Paris-Sud, Centre d'Orsay, 91405 Orsay CEDEX, France
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A scale-invariant theory of resonant Rayleigh scattering by fractal clusters is developed. Our main result is that the scattering cross section is greatly enhanced, because of the presence of very high local fields, which are correlated and strongly fluctuating. Simulations dealing with two examples of fractal structures, namely, random walk and cluster-cluster aggregates, are presented. The numerical results confirm the theoretical predictions of the scaling behavior for both absorption and scattering, and allow us to obtain the corresponding exponents. In addition to the results for scattering, the large scale of the simulations of the present work provides a comprehensive confirmation and a substantial extension of a recent study by Markel, Muratov, Stockman, and George [Phys. Rev. B **43**, 8183 (1991)].

I. INTRODUCTION

Optical properties of fractal clusters[1,2] and in particular scattering of light by fractals[3–13] have recently been an area of active research. It is now well known that the scattered wave bears information about the self-similar geometry of the fractal structure and allows the determination of its fractal dimension D through the formula $I_s(k) \propto k^{-D}$, where k is the wave vector of the incident beam.

However, together with geometrical properties, it is of great interest to study manifestations of the dynamical properties of fractals via light scattering. When individual particles of a fractal cluster possess a high quality ("sharpness") of optical resonance, the light-induced dipole interaction (at driving frequency ω) between polarizable particles can be very strong. Such interaction leads to the formation of collective resonances—eigenmodes—of the system. If the field frequency ω is close to a frequency of an eigenmode of fractal excitations, the scattering has a collective character and is determined both by geometrical and by dynamical properties. In this case light is scattered by collective fractal excitations rather than by individual particles.

As a direct consequence of the scaling properties of the eigenmodes of fractal dipole excitations, the light scattering also exhibits a scale-invariant behavior. We show in this paper that the enhancement factor of the scattering by fractals is characterized by a scaling exponent $d_s = d_0 + 1$ where d_0 is the "optical spectral dimension" already introduced in Ref. 1. This result is confirmed by numerical simulations. The exponents d_s, d_0 are determined for two types of fractals: random walk (RW; with fractal dimension $D = 2$) and cluster-cluster aggregates (CC[14]; with fractal dimensional $D = 1.78$). We also show that, for fractals consisting of particles with a high quality of optical resonance, the resonant Rayleigh scattering is greatly enhanced. This is due to the existence of strongly fluctuating high local fields, significantly exceeding that of the incident beam.[2] Together with resonant enhancement the scattering is also improved by coherence due to the fractality. The huge enhancement for scattering, obtained in our numerical simulations, is in good agreement with theoretical predictions.

The interaction of light-induced dipoles in fractal cluster was considered earlier within the mean-field approach[4] and the binary approximation.[2] However, neither method led to a scale-invariant theory for fractal excitations. Such a theory for the polarizability of small (in comparison with the wavelength) fractal clusters was developed in Ref. 1. In the present numerical simulations we use ten times as many particles (up to $N = 256$) and ten times more clusters (up to $N_c = 1000$) than in Ref. 1. This has allowed us, besides the above-mentioned results on resonant Rayleigh scattering, to obtain a comprehensive confirmation of the main fundamentals of optics of fractals established in Ref. 1.

General consideration of scattering by cluster systems is given in Sec. II. The results obtained are then applied to the scattering by fractal clusters in Sec. III. In Sec. IV we present results of numerical simulations. Final discussion of our results and main conclusions are given in Sec. V.

II. GENERAL EXPRESSIONS

Consider a fractal, consisting of polarizable monomers with interaction between them at the driving frequency ω. The fractal is subjected to an external electrical field, whose value at the site of the ith monomer ($i = 1, \ldots, N$) is equal to $\tilde{\mathbf{E}} = \mathbf{E}^{(0)} \exp(-i\omega t + i\mathbf{k}\cdot\mathbf{r}_i)$. The wavelength λ is supposed to be greater than the size of a monomer but arbitrary in comparison with the size of the cluster. The amplitudes $\mathbf{d}^i$ of the transitional dipole momenta $\tilde{\mathbf{d}}^i = \mathbf{d}^i \exp(-i\omega t + i\mathbf{k}\cdot\mathbf{r}_i)$ obey the system of equations

$$-(X + i\delta) d^i_\alpha = E^{(0)}_\alpha - \sum_j V^{ij}_{\alpha\beta} d^j_\beta \ , \tag{1}$$

where $X=-\mathrm{Re}\chi_0^{-1}$, $\delta=-\mathrm{Im}\chi_0^{-1}$, and χ_0 is the polarizability of an isolated monomer;

$$V_{\alpha\beta}^{ij}=\frac{a^{ij}\delta_{\alpha\beta}-3b^{ij}n_\alpha^{ij}n_\beta^{ij}}{r_{ij}^3}\exp(ikr_{ij}-i\mathbf{k}\cdot\mathbf{r}_{ij}) , \tag{2}$$

$$a^{ij}=1-ikr_{ij}-(kr_{ij})^2 ,$$
$$b^{ij}=1-ikr_{ij}-\tfrac{1}{3}(kr_{ij})^2 , \tag{3}$$

and $\mathbf{r}_{ij}\equiv\mathbf{r}_i-\mathbf{r}_j$. The Greek subscripts in (2) and (3) stand for tensor components (summation over repeated indices is implied). The interaction (2) and (3) includes the near-zone (nonradiative) transitional and far-zone (radiative) terms of the dipole field.

The fractal cluster produces a scattered wave whose asymptotic form, involving a vector scattering amplitude $\mathbf{f}(\mathbf{k}',\mathbf{k})$, is

$$E_\alpha^s=\frac{\exp(ikr)}{r}E^{(0)}f_\alpha(\mathbf{k}',\mathbf{k}) , \tag{4}$$

$$f_\alpha(\mathbf{k}',\mathbf{k})=k^2(E^{(0)})^{-1}\sum_i\tau_{\alpha\beta}d_\beta^i\exp(i\mathbf{k}\cdot\mathbf{r}_i-i\mathbf{k}'\cdot\mathbf{r}_i) , \tag{5}$$

$$\tau_{\alpha\beta}=\delta_{\alpha\beta}-s_\alpha s_\beta;\quad \mathbf{s}\equiv\mathbf{k}'/k , \tag{6}$$

where $\mathbf{k}'$ is the wave vector of the scattered wave. To obtain (4)–(6) we use the approximate equality $k|\mathbf{r}-\mathbf{r}_i|\simeq kr-\mathbf{k}'\cdot\mathbf{r}_i$, which is valid for distances much greater than the size of the cluster. The set of Eqs. (1)–(6) is the basis for calculations of optical properties of clusters.

We seek to calculate the ensemble averages of the scattering cross section

$$\sigma_s=\langle\int d\Omega_s|f(\mathbf{k}',\mathbf{k})|^2\rangle \tag{7}$$

(where Ω_s is the direction of $\mathbf{k}'$), the extinction cross section

$$\sigma_e=\langle 4\pi k\sum_i\mathrm{Im}\chi_{\alpha\beta}^i e_\alpha e_\beta\rangle , \tag{8}$$

and the absorption cross section

$$\sigma_a=\sigma_e-\sigma_s . \tag{9}$$

Vector $\mathbf{e}$ in (8) is the unit one, $\mathbf{e}\equiv\mathbf{E}^{(0)}/E^{(0)}$; angle brackets $\langle\cdots\rangle$ denote the average over an ensemble of clusters. The polarizability of the ith monomer $\chi_{\alpha\beta}^i$ in (8) is defined by

$$d_\alpha^i=\chi_{\alpha\beta}^i E_\beta^{(0)} . \tag{10}$$

Inserting (5) and (10) into (7) and performing the integration we obtain

$$\sigma_s=4\pi k^4N^2\langle\phi_{\alpha\alpha'}^{ii'}\chi_{\alpha\beta}^i\chi_{\alpha'\beta'}^{i'*}e_\beta e_{\beta'}\rangle , \tag{11}$$

where

$$\phi_{\alpha\alpha'}^{ii'}=\cos\mathbf{k}\cdot\mathbf{r}_{ii'}(\phi_1\delta_{\alpha\alpha'}-\phi_2 n_\alpha^{ii'}n_{\alpha'}^{ii'}) , \tag{12}$$

$$\phi_1=\frac{\sin kr_{ii'}}{kr_{ii'}}+\frac{\cos kr_{ii'}}{(kr_{ii'})^2}-\frac{(\sin)kr_{ii'}}{(kr_{ii'})^3} , \tag{13}$$

$$\phi_2=\frac{\sin kr_{ii'}}{kr_{ii'}}+3\frac{\cos kr_{ii'}}{(kr_{ii'})^2}-3\frac{\sin kr_{ii'}}{(kr_{ii'})^3} . \tag{14}$$

Let us now average (11) over the orientations of a cluster as a whole (denoted below as $\langle\cdots\rangle_0$). In order to fulfill such averaging analytically let us use the "unlinking procedure"

$$\langle\phi_{\alpha\alpha'}^{ii'}\chi_{\alpha\beta}^i\chi_{\alpha'\beta'}^{i'*}\rangle_0\to\langle\phi_{\alpha\alpha'}^{ii'}\rangle_0\langle\chi_{\alpha\beta}^i\chi_{\alpha'\beta'}^{i'*}\rangle_0 . \tag{15}$$

The procedure (15) is valid usually in the case of an isotropic system. For fractal clusters having strong fluctuations of its shape, the validity of (15) is not evident. Validity of such averaging, however, is strongly supported by our numerical simulation (see Sec. IV).

Averaging over orientations in (11) with using (15) gives after some elementary transformations

$$\sigma_s=\frac{2\pi}{15}k^4N^2[\langle(15\phi_1^2-10\phi_1\phi_2+2\phi_2^2)\mathrm{Tr}(\hat{\chi}^i\hat{\chi}^{i'*})\rangle$$
$$+\langle\phi_2^2\mathrm{Tr}\hat{\chi}^i\mathrm{Tr}\hat{\chi}^{i'*}\rangle] , \tag{16}$$

where $\hat{\chi}\equiv\chi_{\alpha\beta}$, $\mathrm{Tr}\,A\equiv A_{\alpha\alpha}$. Note that the isotropy of the cluster has eliminated the dependence on the initial polarization $\mathbf{e}$.

Averaging over the orientations of the extinction cross section in (8) gives

$$\sigma_e=\frac{4\pi}{3}kN\langle\mathrm{Im}\chi_{\alpha\alpha}^i\rangle . \tag{17}$$

Let us introduce the factor F_s by which the scattering per monomer is enhanced

$$F_s=\frac{\sigma_s}{N\sigma_s^{(0)}} , \tag{18}$$

where $\sigma_s^{(0)}$ is the single-particle scattering cross section $[(\chi_0)_{\alpha\beta}\equiv\chi_0\delta_{\alpha\beta}]$:

$$\sigma_s^{(0)}=\frac{8\pi}{3}k^4|\chi_0|^2 . \tag{19}$$

When the size of the cluster is much less than the wavelength, the values $\phi_{1,2}$ in (16) do not depend on the statistics of clusters ($\phi_1\simeq\frac{2}{3}$, $\phi_2\simeq 0$) and the enhancement factor of the scattering is

$$F_s=\tfrac{1}{3}N|\chi_0|^{-2}\langle\mathrm{Tr}(\hat{\chi}^i\hat{\chi}^{i'*})\rangle\quad(kR_c\ll 1) . \tag{20}$$

Far from the collective resonances of the cluster (nonresonant case) the interaction between monomers is negligible because the collective eigenmodes are not excited, and we can take $\chi_{\alpha\beta}^i=\chi_0\delta_{\alpha\beta}$. Such an expression for χ^i corresponds to neglecting the interaction term in (1) compared with X. For small clusters we obtain in this way from (20) the trivial limit

$$F_s^{(NR)}=N\quad(kR_c\ll 1) . \tag{21}$$

Note that the substitution of $\chi_{\alpha\beta}^i=\chi_0\delta_{\alpha\beta}$ into (17) gives only an absorption part $\sigma_a=4\pi kN\,\mathrm{Im}\chi_0$ of the total cross section σ_e and does not take into account the contribution of the scattering into the extinction. Since scattering depends on both (real and imaginary) parts of the polarizability: $\sigma_s\propto\chi\chi^*$, one can put $\chi_{\alpha\beta}^i=\chi_0\delta_{\alpha\beta}$ in the formula for σ_s even if only $X=-\mathrm{Re}\chi_0^{-1}$ (and not both X and $\delta=-\mathrm{Im}\chi_0^{-1}$) is much larger than the contri-

bution of the interaction in (1) (the value of δ for this can be arbitrary small comparing with the interaction). However, in order to find the contribution of the scattering into the extinction (which is determined only by imaginary part of polarizability: $\sigma_e \propto \mathrm{Im}\chi$) we should retain corrective term in χ^i which takes into account the interaction between particles (even if this term is much smaller than X).

Let us emphasize that all the results which have been presented in this section are quite general and valid for any cluster (fractal or not). We will now focus on the more particular case of fractal clusters.

III. LIGHT SCATTERING IN THE COLLECTIVE REGION OF FRACTAL EXCITATIONS

The number of particles in a fractal cluster of size R_c (R_c is a radius of gyration) and the pair correlation function are described, respectively, by the following scaling dependencies:

$$N=(R_c/R_0)^D ,$$
$$p(r)=\frac{D}{4\pi}R_c^{-D}r^{D-3} , \tag{22}$$
$$\langle p(r)\rangle=4\pi\int_0^{R_c} r^2 p(r)dr=1 .$$

Let us now reduce the system of Eqs. (1)–(3). The last term in (1) gives the contribution of all monomers to the local field acting on the ith monomer. To estimate the contribution of monomers located at large distances $r_{ij}>k^{-1}$ from a given ith monomer we can use the mean-field approach[4] which gives, using (22)

$$|v|\sim\begin{cases} R_0^{-3}(kR_0)^{3-D}, & \text{for } (D<2) , \\ R_0^{-3}(kR_0)N^{1-2/D}, & \text{for } (D>2) , \end{cases} \tag{23}$$

where

$$v\equiv\left\langle \sum_{\substack{j \\ (r_{ij}>k^{-1})}} V_{\alpha\beta}^{ij}e_\alpha e_\beta \right\rangle . \tag{24}$$

When the spectral variable X in (1) satisfies the condition

$$|X|\gg|v| , \tag{25}$$

we can neglect the contribution to d^i of the region $r_{ij}>k^{-1}$ and reduce (1)–(3) to the system

$$-(X+i\delta)d_\alpha^i=E_\alpha^{(0)}-\sum_j W_{\alpha\beta}^{ij}d_\beta^i , \tag{26}$$

where

$$W_{\alpha\beta}^{ij}=\begin{cases}(\delta_{\alpha\beta}-3n_\alpha^{ij}n_\beta^{ij})r_{ij}^{-3}, & \text{if } kr_{ij}<1 \quad (i\neq j) ; \\ 0, & \text{otherwise} . \end{cases} \tag{27}$$

For resonant conditions $X=\Delta_i$ (Δ_i is the shift of the resonance due to the interaction W) a condition of such a reduction is more strict

$$\frac{|v|}{\delta}<1 . \tag{28}$$

The reduction of (1)–(3) to (26) and (27) physically means that the contribution of far-zone fields of the monomers to the dipole d^i of the ith monomer is negligible and, thus, multiple scattering can be neglected. After such a reduction the interaction (27) together with a spatial scaling leads to the scaling of optical properties of fractals. Thus, the condition (25) for the spectral variable X is a necessary condition to obtain the scaling of optical excitations of fractals.

Note also that the mean-field approach is not applicable for the description of interaction at small distances (less than a wavelength).[2] This approach gives either a divergence at small distances as r^{D-3} (without averaging over the orientations) or exactly zero after averaging over the orientations, and therefore strong fluctuations, which are critical for optical properties of fractals, cannot be taken into account.[1,2]

Following Ref. 1, we express the solution of the system (26) in terms of the eigenstates of the Hermitian operator (27) [$\hat{W}|n\rangle=w_n|n\rangle$]:

$$\chi_{\alpha\beta}^i=\sum_{n,j}\Lambda_n(i\alpha|n)(n|j\beta) , \tag{29}$$
$$\Lambda_n=(\chi_0^{-1}+w_n)^{-1} . \tag{30}$$

From the form of the interaction operator (27) and the solution (29), the exact sum rules follow:

$$\frac{1}{\pi}\int_{-\infty}^{\infty}\mathrm{Im}\chi_{\alpha\beta}^i(X)dX=\delta_{\alpha\beta}, \quad \int_{-\infty}^{\infty}X\,\mathrm{Im}\chi(X)dX=0 . \tag{31}$$

The quantity $\mathrm{Im}\chi(X)$ ($\chi\equiv\frac{1}{3}\langle\chi_{\alpha\alpha}\rangle$) has a symmetrical power-law dependence in the scaling region

$$\mathrm{Im}\chi(X)\sim R_0^3(R_0^3|X|)^{d_0-1} , \tag{32}$$

where $d_0(0<d_0<1)$, is the optical spectral dimension,[1] which is very important in the description of optical properties of fractals.

From the sum rules (31) and the power-law dependence of $\mathrm{Im}\chi(X)$ it follows that the contribution of nondiagonal terms in (29) with $i\neq j$, after averaging over the ensemble of clusters, is negligible. Thus, for $|X|\gg\delta$, we can express the quantity $\mathrm{Im}\chi(X)$ in terms of the density of eigenstates $\nu(X)$:[1]

$$\mathrm{Im}\chi(X)=\frac{\pi}{3}\nu(X) , \tag{33}$$

where $\nu(X)$ is defined in the usual way by

$$\nu(X)=\frac{1}{N}\left\langle\sum_n\delta(X-w_n)\right\rangle .$$

It is worth noticing, as follows from (32) and (33), that the dimension d_0 plays the same role in the case of dipolar fractal excitations as the fracton dimension $\tilde{d}$ (Ref. 15) in the case of vibrational excitations. However, these indices d_0, $\tilde{d}$, and the corresponding dispersion relations (see below) are different for vibrational (Goldstone-type) and dipolar (non-Goldstone-type) fractal excitations.[16]

By analogy with the nonresonant case [see the discussion after formula (21)] substitution of the approximate expression (29) for χ^i into (17) gives only the absorption

part σ_a of the total cross section σ_e [see also (62) and the text followed after (62)]. Taking this into account and using (33) we find

$$\sigma_a \sim kN\nu(X) , \tag{34}$$

and together with (32) and (33)

$$\sigma_a \sim kNR_0^3 (R_0^3|X|)^{d_0-1} . \tag{35}$$

The scaling-transformation properties of X can be found from the condition of invariance of the absorption (35) with respect to the change of the minimal size (spatial resolution) R_0.[1] After substituting $N=(R_c/R_0)^D$ into (35) this condition gives

$$|X| \propto R_0^{-(3d_0-D)/(d_0-1)} . \tag{36}$$

One can construct only a single invariant of the scaling transformation based on (36) which contains both X and R_0 and which has the dimensionality of length

$$L_X \sim R_0 (R_0^3|X|)^{(d_0-1)/(3-D)} . \tag{37}$$

The quantity L_X in (37) plays the role of a coherence length and characterizes the length of localization of the eigenstates of the fractal excitations with eigenvalues $w_n = X$.[1,16]

Scaling, i.e., the invariance with respect to the change of the minimal scale R_0, can only take place under the condition

$$R_0 \ll L_X \ll \lambda . \tag{38}$$

Note that in accordance with (37), the requirement $L_X \ll \lambda$ is automatically fulfilled by the condition (25). Taking into account (23), (25), and (37) we find the scaling region for large clusters ($kR_c \gg 1$) in terms of the spectral variable X

$$(kR_0)^{3-D} \ll R_0^3|X| \ll 1 \quad (D<2) , \tag{39}$$

$$kR_0 N^{1-2/D} \ll R_0^3|X| \ll 1 \quad (D>2) . \tag{40}$$

The condition (40) is fulfilled at large values kR_c only if

$$1 \ll kR_c \ll (kR_0)^{2/(2-D)} \quad (D>2) , \tag{41}$$

which is assumed to be the case. Besides (39) and (40), the necessary condition for scaling is $|X| \gg \delta$, which is compatible with (39) and (40) when the quality factor Q of the monomer's resonance is high enough.

In the case of small clusters ($R_c \ll k^{-1}$) the scaling region is defined from the condition $R_0 \ll L_X \ll R_c$. Using (37) we obtain[1]

$$N^{-(3/D-1)/(1-d_0)} \ll R_0^3|X| \ll 1 \quad (kR_c \ll 1) .$$

In a given spectral interval $(X, X+dX)$ there are $\sim N\nu(X)dX$ eigenmodes excited. These excitations are centered in different points of the fractals and have [at conditions (39) and (40)] the localization length L_X much less than the wavelength λ. Taking this into account we can average $\phi^{ii'}$ and $\chi^i\chi^{i'*}$ in (16) over interparticle distances r_{ij} independently. After averaging of the functions $\phi_{1,2}$ in (16) we find

$$\sigma_s = \frac{2\pi}{15} k^4 N (K_1 \langle \mathrm{Tr}(\hat{\chi}^i \hat{\chi}^{i'*}) \rangle + K_2 \langle \mathrm{Tr}\hat{\chi}^i \mathrm{Tr}\hat{\chi}^{i'*} \rangle) , \tag{42}$$

$$K_1 = \begin{cases} D\Gamma(D-1)2^{1-D}\cos\dfrac{\pi}{2}(D-2)(kR_0)^{-D}\left[\dfrac{7}{2-D}-\dfrac{2}{4-D}+\dfrac{2}{6-D}\right] , & \text{for } (D<2), \\ \dfrac{7}{2}(kR_0)^{-2}\ln((kR_0)^2N) , & \text{for } (D=2) , \\ \dfrac{7}{2}\dfrac{D}{D-2}(kR_0)^{-2}N^{1-D/2} , & \text{for } (D>2) , \end{cases} \tag{43}$$

$$K_2 = \begin{cases} D\Gamma(D-1)2^{1-D}\cos\dfrac{\pi}{2}(D-1)(kR_0)^{-D}\left[\dfrac{1}{2-D}-\dfrac{6}{4-D}+\dfrac{6}{6-D}\right] , & \text{for}(D<2) , \\ \dfrac{1}{2}(kR_0)^{-2}\ln((kR_0)^2N) , & \text{for } (D=2) , \\ \dfrac{1}{2}\dfrac{D}{D-2}(kR_0)^{-2}N^{1-2/D} , & \text{for}(D>2) , \end{cases} \tag{44}$$

where $\Gamma(\cdots)$ is the γ function. The enhancement factor of scattering F_s follows from (42), (18), and (19)

$$F_s = \tfrac{1}{20}|\chi_0|^{-2}(K_1 \langle \mathrm{Tr}(\hat{\chi}^i \hat{\chi}^{i'*}) \rangle + K_2 \langle \mathrm{Tr}\hat{\chi}^i \mathrm{Tr}\hat{\chi}^{i'*} \rangle) . \tag{45}$$

In the limit of nonresonant scattering (see above) (45) reduces to the simple formula

$$F_s = \tfrac{3}{20}(K_1 + 3K_2) , \tag{46}$$

which is in exact agreement with results obtained by Berry and Percival.[4]

Substituting (29) into (45) and expanding the product of factors Λ in simple factors

$$\Lambda_n\Lambda^*_{n'}=-\frac{1}{(w_n-w_{n'})-2i\delta}\{\Lambda_n-\Lambda^*_{n'}\}\ ,$$

we find

$$F_s=-\tfrac{1}{20}|\chi_0|^{-2}\Big\langle\sum_{n,n',j,j'}\frac{1}{(w_n-w_{n'})-2i\delta}(\Lambda_n-\Lambda^*_{n'})[K_1(i\alpha|n)(n|j\beta)(i'\beta|n')(n'|j'\alpha)$$
$$+K_2(i\alpha|n)(n|j\alpha)(i'\beta|n')(n'|j'\beta)]\Big\rangle\ . \tag{47}$$

The minimal difference Δ_n between eigenvalues w_n and $w_{n'}$ in (47) can be estimated as

$$\Delta_n\sim\nu^{-1}\sim R_0^{-3}(R_0^3|X|)^{1-d_0}\ . \tag{48}$$

Using (33) one can see that at the condition

$$R_0^3|X|\gg(R_0^3\delta)^{1/(1-d_0)} \tag{49}$$

the mode spacing Δ_n is always much larger than δ. Thus, for monomers with high optical-resonance quality (small value of δ), when (49) is the case, one can neglect in (47) the contribution of terms with $n\neq n'$. This ultimately gives

$$F_s=\frac{1}{5}\gamma\frac{|\chi_0|^{-2}}{|\mathrm{Im}\chi_0^{-1}|}\Phi(X,R_0)\ , \tag{50}$$

where

$$\gamma=\begin{cases}D\Gamma(D-1)2^{2-D}\cos\dfrac{\pi}{2}(D-1)(kR_0)^{-D}\left[\dfrac{1}{2-D}-\dfrac{1}{4-D}+\dfrac{1}{6-D}\right], & \text{for } (D<2)\ ,\\ (kR_0)^{-2}\ln[(kR_0)^2N], & \text{for } (D=2)\ ,\\ \dfrac{D}{D-2}(kR_0)^{-2}N^{1-2/D}, & \text{for } (D>2)\ ,\end{cases} \tag{51}$$

and

$$\Phi(X,R_0)=\Big\langle\sum_{n,j,j'}\mathrm{Im}\Lambda_n(i\alpha|n)(n|j\beta)(i'\beta|n)(n|j'\alpha)\Big\rangle\ . \tag{52}$$

It follows from (52) that the function $\Phi(X,R_0)$ satisfies the sum rule

$$\int_{-\infty}^{\infty}\Phi(X)X\,dX=0\ . \tag{53}$$

Therefore, in the scaling regions (39) and (40) $\Phi(X,R_0)$ should have a symmetrical power-law dependence with coefficient which can be defined from dimensionality arguments

$$\Phi(X,R_0)\sim R_0^3(R_0^3|X|)^{\Theta}\ , \tag{54}$$

where Θ is a corresponding exponent.

In accordance with (18), (19), and (50), the function $\Phi(X,R_0)$ must have the same dependence on X as σ_s: $\Phi(X)\propto F_s|\chi_0|^2\propto\sigma_s(X)$. Taking into account (9) one can conclude that all three quantities σ_e, σ_s, and σ_a should have the same exponent in the scaling region. It follows from (35) that this index is d_0-1. Thus, we have

$$\Theta=d_0-1\ . \tag{55}$$

Using (20), (54), (55), and the relation [see (45)–(50)]

$$\langle\mathrm{Tr}(\hat\chi^i\hat\chi^{i'*})\rangle\sim|\mathrm{Im}\chi_0^{-1}|^{-1}\Phi(X,R_0)\ ,$$

one can find the scattering enhancement in the case of small clusters ($kR_c\ll1$):

$$F_s\sim\frac{N}{R_0^3|\mathrm{Im}\chi_0^{-1}|}(R_0^3|X|)^{d_s}\quad(kR_c\ll1)\ , \tag{56}$$

where

$$d_s=d_0+1\ . \tag{57}$$

Equation (56) was obtained by substituting $|\chi_0|^{-2}\simeq|X|^2$ into (20), which is valid for $|X|\gg\delta$.

For large clusters ($kR_c\gg1$) the limiting behavior, as follows from (50), (51) and (54), (55), depends on D:

$$F_s\sim\frac{(kR_0)^{-D}}{R_0^3|\mathrm{Im}\chi_0^{-1}|}(R_0^3|X|)^{d_s}\quad(D<2)\ , \tag{58}$$

$$F_s\sim\frac{(kR_0)^{-2}}{R_0^3|\mathrm{Im}\chi_0^{-1}|}\ln((kR_0)^2N)(R_0^3|X|)^{d_s}\quad(D=2)\ , \tag{59}$$

$$F_s\sim\frac{(kR_0)^{-2}}{R_0^3|\mathrm{Im}\chi_0^{-1}|}N^{1-2/D}(R_0^3|X|)^{d_s}\quad(D>2)\ , \tag{60}$$

where d_s is given by (57).

In accordance with (37), increasing $|X|$ leads to an improved localization of fractal dipole excitations and as a result, to an increase in the intensities of local fields inside the fractal. High local fields (significantly exceeding the exciting external field) ultimately lead to an enhanced scattering. Thus, the scattering enhancement factor F_s should increase with increasing $|X|$ (the exponent d_s is positive). It is worth noticing that localization of dipole optical excitations takes place only on fractal structure and disappears in the limit of a conventional medium where $D=3$.

Let us now use the mean-field approach in order to take into account the contribution of the region $r_{ij}>k^{-1}$ [neglected in (26)] into d^i. This approach allows us to find the improved formula for the polarizability $\tilde{\chi}_i$:

$$\tilde{\chi}_i \sim \frac{\chi_i}{1+v\chi_i}=\frac{\chi_i+v^*|\chi_i|^2}{|1+v\chi_i|^2}\ , \tag{61}$$

where $\chi_i \equiv 1/3\chi^i_{\alpha\alpha}$ is determined by (29). Within this approximation, multiple scattering can be neglected provided $|v\chi_i| \ll 1$. If this is the case, a substitution of $\tilde{\chi}_i$ into (17) gives

$$\sigma_e \sim kN\ \mathrm{Im}\{\langle\chi_i\rangle+v^*\langle|\chi_i|^2\rangle\}\ . \tag{62}$$

One can make sure that the second term in (62) is of the order of σ_s determined in (18), (19), and (58)–(60). Thus, the first term in (62) corresponds to the absorption part σ_a in the total cross section σ_e in accordance with (32) and (35). Substitution of $\tilde{\chi}_i$ into (45) results of course, in the same formulas (58)–(60) for F_s in the limit of single scattering.

IV. NUMERICAL SIMULATIONS

A numerical solution of the problem of optical excitation of the ensemble of clusters, consisting of N interacting particles, is restricted to $N\sim 10^3$ or even less (for reasonable computing time) due to the order of the matrix to be diagonalized and the numbers of eigenfunctions to be determined. The region of spatial scaling ($R_0 \ll r \ll R_0 N^{1/D}$) for N of order of a few hundred is rather short and not always well pronounced. Accordingly, it is difficult to obtain the dynamical scaling. To resolve this difficulty a model of a "diluted" fractal can be used fruitfully.[1] In this model each monomer of the initial fractal (consisting of the large enough number of particles) remains in its place (with probability $\beta \ll 1$) or is removed from the fractal (with probability $1-\beta$). The fractal as a whole is then reduced in size by a factor $(1/\beta)^{1/D}$. The resulting fractal turns out to have the same scaling properties as the initial one. The power-law behavior of the correlation function $g(r) \propto r^{D-3}$ holds for this fractal (with the same exponent D as the initial one) down to smaller distances $r_0=\beta^{1/D}R_0 \ll R_0$. It is important that the dilution (random mass decimation) does not affect the properties in the region of collective excitations. Since collective excitations have a large coherence length L, they are not sensitive to the small-scale details of the fractal structure and therefore should be the same for the original and diluted fractals.

We study two types of fractals: random walks [RW: $D=2$; $R_0=R_1/(6)^{1/2}$], and cluster-cluster aggregates [CC (Ref. 14): $D\simeq 1.78$; $R_0 \simeq R_1/3$; where R_1 is the lattice period). All clusters used here were subjected to 32-fold decimation ($\beta \simeq 0.03$) except in simulations done with β as small as 10^{-3} for comparison (see Fig. 5). Decreasing the value of β, as shown in the simulation does not affect the results. The calculated values are averaged over a large ensemble of fractals (about 10^3).

The polarizability was computed from (29) by diagonalizing the matrix (27) using the numerical QL method.[17]

In accordance with (32) and (35) the absorption of a cluster per monomer is determined by the imaginary part of the polarizability $\mathrm{Im}\chi \equiv \frac{1}{3}\langle \mathrm{Im}\chi_{\alpha\alpha}\rangle$. The quantity $\mathrm{Im}\chi$, plotted as a function of X, is shown in Fig. 1 for two different numbers of particles: $N=128$ and $N=256$ and for $\delta=0.001$. Here and in all other figures we put $R_0=1$. From Fig. 1 it is clear that there is no change with the number of particles. The absorption is symmetrical in X and described by a resonant contour in accordance with (32). The width of the fractal absorption greatly exceeds that of an individual monomer ($\sim\delta$). A double-logarithmic plot of $X\ \mathrm{Im}\chi$ versus X is shown in Fig. 2(a) in the range $X=0.002$ to 0.6. The last value of X is 0.6 [but not 1 as follows from (39)] because for $X>0.6$ fluctuations become of the same order of magnitude as the mean value (more precisely, the upper X value, here and in all the following, is chosen such that fluctuations become equal to twice the mean). One can see from Fig. 2(a) that absorption for the RW model is well described by a power-law dependence on X and the slope gives the optical spectral dimension $d_0=0.4\pm0.1$. The straight line in the figure corresponds to $d_0=0.38$ as found by a least-squares method. This result is in perfect agreement with Ref. 1.

For the CC model the scaling behavior is also observed [Figs. 3 and 4(a)]. We have obtained the value of the optical spectral dimension for the cluster-cluster aggregates

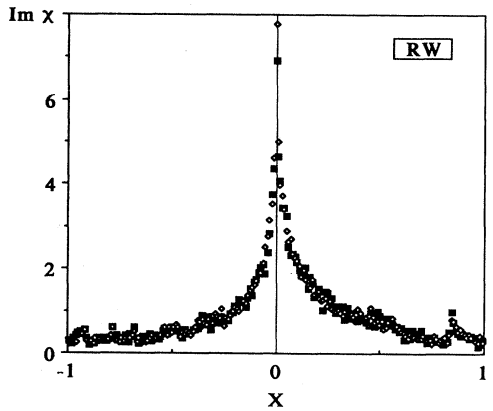

FIG. 1. Plot of $\mathrm{Im}\chi$ as a function of X for the random walk model with $\delta=0.001$. Symbols ■ and ◇ refer to $N=128$ and $N=256$, respectively.

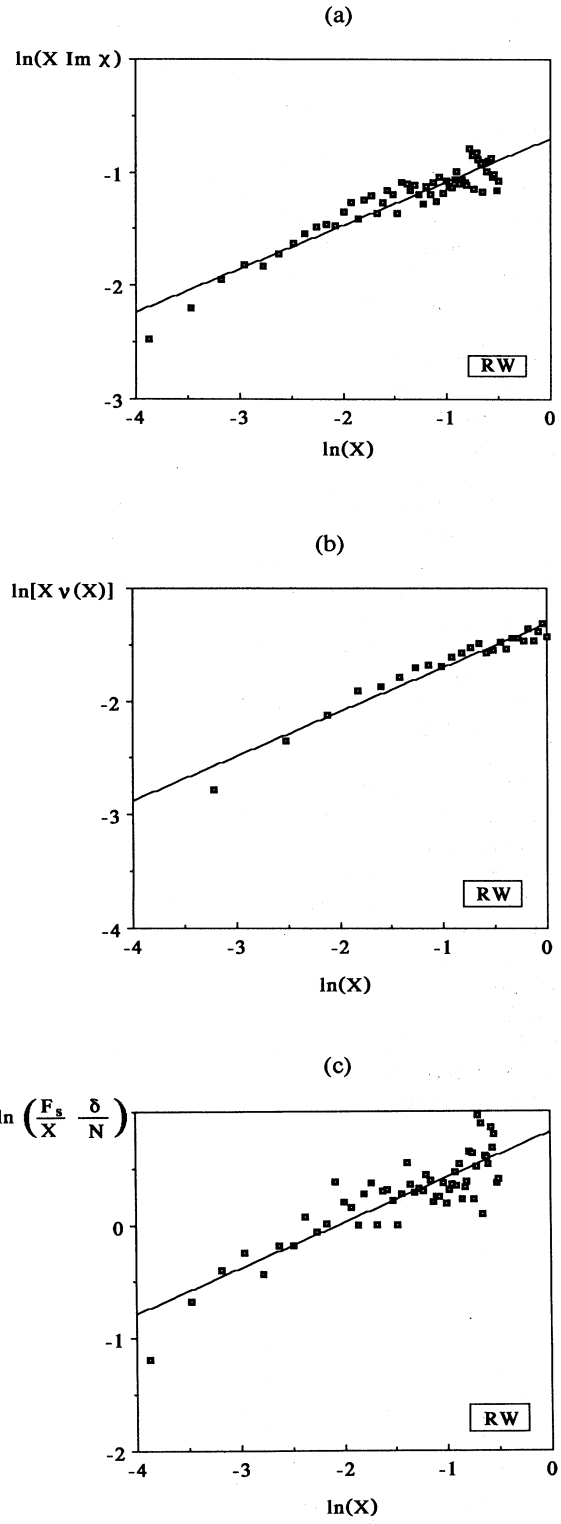

FIG. 2. (a) ln-ln plot of Imχ multiplied by X as a function of X. The slope of $d_0=0.38$ is indicated. (b) ln-ln plot of the density of eigenstates $\nu(X)$ multiplied by X as a function of X. The slope of $d_0=0.39$ is indicated. (c) ln-ln plot of the scattering enhancement F_s multiplied by $1/N\ \delta/X$ as a function of X. $N=128$, $\delta=0.001$. The slope is $d_0=0.40$.

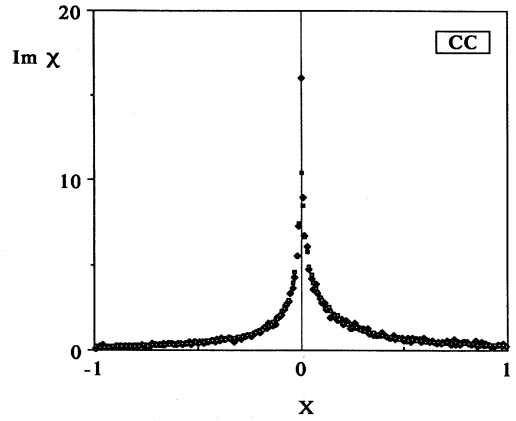

FIG. 3. Plot of Imχ as a function of X for cluster-cluster aggregates. Symbols ♦ and □ refer to $\delta=0.001$ and $\delta=0.005$, respectively.

$d_0=0.3\pm0.1$ (the slope $d_0=0.28$ is shown in the figure). From Fig. 3 one can also see that, in accordance with the theory, there is no dependence of the absorption on δ for $|X|\gg\delta$. Other quantitative differences between the two models can be seen by comparing Fig. 1 and Fig. 3, which correspond to the same set of parameters (except for the number of particles which is $N=64$ for the CC model).

Thus, we can conclude, the absorption has a universal scaling behavior in the region of collective fractal excitations in agreement with the theory.

According to formula (33) the density of eigenstates must show the same scaling behavior. Figures 2(b) and 4(b) support this result and give the same slope as in the case of the absorption: $d_0=0.4\pm0.1$ for the RW model (in agreement with Ref. 1) and $d_0=0.3\pm0.1$ for the CC model.

We also present in Fig. 5 the density of eigenstates for two very different values of β: $\beta=32^{-1}$ and $\beta=1000^{-1}$ ($N=30$). One can see that for $\beta=32^{-1}$ the overall behavior is quite similar to an order of magnitude (although the results are more noisy). This proves that the dilution does not affect collective fractal properties. The order of magnitude of $\nu(X)$ we obtain is close to that of the absorption (Fig. 1) in the scaling region, according to the theory. The small asymmetry of the density of eigenstates which takes place in the central part outside the scaling region is a finite-size effect.

We also numerically calculated the corrective term (24) neglected in our theoretical approach when using the reduction procedure [see (23)–(27)]. This correction did not exceed 10%.

The enhancement factor of the scattering F_s was computed from (18) using the general formulas (11)–(14) for σ_s. The factor $F_s(R_0^3\delta)/N$ ($R_0=1$) is plotted in Fig. 6 as a function of X for two different size clusters ($N=128$ and $N=256$). Note that the number of particles and value of kR_0 ($kR_0=0.1$) that we used here correspond to intermediate cluster size: $kR_c\sim1$, because $kR_0=0.1$ for $D=2$ and $N=256$ gives $kR_c=1.6$ and $kr_{\max}=4.5$ for the

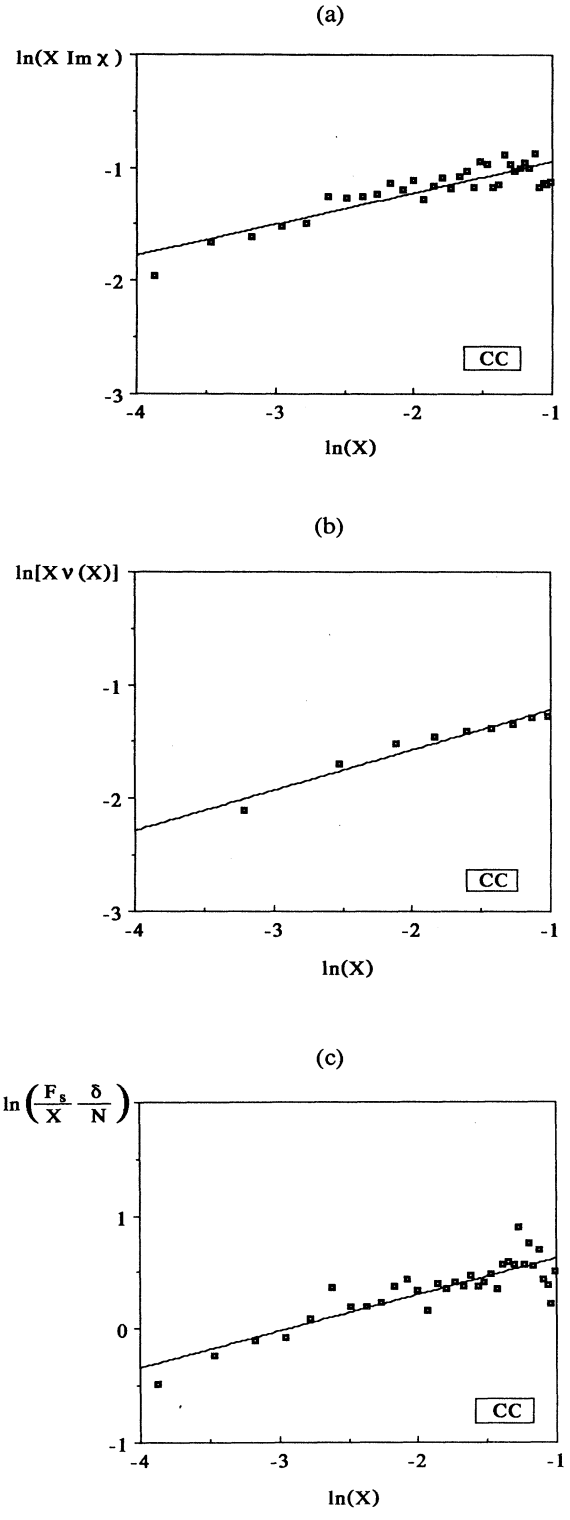

FIG. 4. (a) ln-ln plot of Imχ multiplied by X as a function of X. The slope $d_0=0.28$ is indicated. (b) ln-ln plot of the fractal density of eigenstates $\nu(X)$ multiplied by X. The slope $d_0=0.35$ is indicated. (c) ln-ln plot of the scattering enhancement F_s multiplied by $1/N$ δ/X as a function of X with $N=64$, $\delta=0.001$. The slope $d_0=0.32$ is indicated.

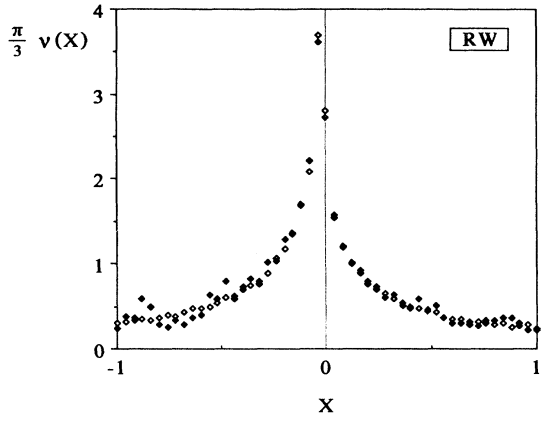

FIG. 5. Plot of the density of fractal eigenstates multiplied by $\pi/3$ as a function of X for the random walk model. Symbols ♦ and ◇ refer to different dilutions ♦:$\beta=0.03$ and ◇:$\beta=0.001$, respectively.

mean value of the largest distance between particles. In accordance with the theory, fluctuations increase with increasing X, as one can see in Fig. 6. The quantities F_s/X also exhibit scaling behavior with power indexes $d_0=d_s-1=0.4\pm0.1$ for RW clusters [Fig. 2(c)] and $d_0=d_s-1=0.3\pm0.1$ for CC aggregates [Fig. 4(c)] in quite good agreement with the values d_0 found from the absorption [Figs. 2(a) and 4(a)] and from the density of states [Figs. 2(b) and 4(b)] in accordance with (57). In contrast to the absorption, the scattering turns out to be enhanced. The value of the enhancement factor obtained here is $F_s\sim10^5$, i.e., roughly equal to the product of the factor $(kR_0)^{-D}$ ($\sim N$ in our case) and the quality of the optical resonance $|R_0^3\mathrm{Im}\chi_0^{-1}|^{-1}$ in agreement with the theoretical formulas (56) and (58).

Note also, that a good agreement of the results of nu-

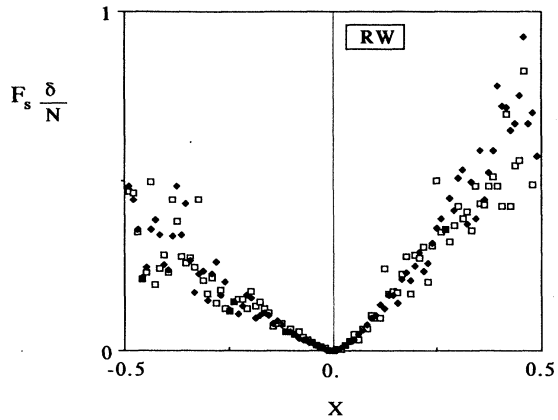

FIG. 6. Plot of the scattering enhancement F_s multiplied by $R_0^3\delta/N$ ($R_0=1$) as a function of X for the random walk model with $\delta=0.001$. Symbols ♦ and □ refer to $N=128$ and $N=256$, respectively.

merical simulation with theoretical formulas obtained, strongly supports the possibility of independent averaging used to get (16) and (42).

V. CONCLUDING REMARKS

We briefly summarize the basic physical results of this paper. Resonant Rayleigh scattering by fractal clusters has been theoretically described. When the frequency of the external field is close to the frequency of the eigenmodes of fractal dipole excitations the scattering occurs on collective excitations rather than on individual particles due to the appearance of strong interaction between particles. The dynamical excitations of the fractal are localized and possess a scaling dependence on the spectral variable $X=-\mathrm{Re}\chi_0^{-1}$ in the scaling region determined by (39) and (40). The corresponding exponent is d_0-1 for the scattering cross section (the same exponent as for absorption cross section) and d_0+1 for the enhancement factor F_s.

It follows from the theory, that the scattering is enhanced by coherence due to the fractality [with a factor $(kR_0)^{-D}$ if $D<2$ and $(kR_0)^{-2}N^{1-2/D}$ if $D>2$; see (58) and (60)]. In addition, the value of the enhancement is proportional to a large factor and, namely, to the quality of optical resonance. The physical reason for such enhancement is the existence of high local fields within a fractal. The external field excites eigenmodes of the fractal with eigenvalues $w_n=X(\omega)$. Local fields E_l of the resonant modes significantly exceed the external one E^0: $Q=E_l/E^0\sim R_0^3|\mathrm{Im}\chi_0^{-1}|^{-1}$. However, the fraction of resonant monomers is small $\sim Q^{-1}$ and therefore, the absorption is not ultimately enhanced. In contrast to absorption the scattering is proportional to the second power of the local field, that is to Q^2. Multiplying this factor by the fraction of resonant monomers Q^{-1}, we obtain the enhancement factor $F_s\propto Q\propto|\mathrm{Im}\chi_0^{-1}|^{-1}$ in agreement with (56) and (58)–(60). It is also interesting to note that the resonant scattering can be significant (comparable with the absorption) even for clusters of size much less than a wavelength [see (18), (19), (35) and (56)].

For isolated particles optical resonance takes place only at $X=-\mathrm{Re}\chi_0^{-1}=0$. The interaction between particles in a fractal leads to the appearance of a set of collective resonances in a wide spectral region. The appearance of additional resonances in another spectral region can lead to the additional enhancement of the scattering due to the improvement of the quality of optical resonance of eigenmodes of the fractal in comparison with the quality of resonance of an isolated monomer. For example, in the case of fractal clusters consisting of small metallic balls, there are collective resonances in the longer wavelengths of the spectrum in comparison with a plasmon resonance of an isolated particle. The monomer's polarizability in this case is $\chi_0=R_m^3(\epsilon-1)/(\epsilon+2)$ (where R_m is the radius of monomer and $\epsilon\equiv\epsilon'+i\epsilon''$ is the permeability of metal) and the corresponding quality of resonance $Q\propto|\mathrm{Im}\chi_0^{-1}|^{-1}\propto|\epsilon-1|^2/\epsilon''$ increases at the transition to the longer wavelengths of the spectrum (the "luster" effect[18]).

To complement the theory we carried out a numerical simulation of the basic equations, including averaging over a large ensemble of clusters. The numerical results obtained confirm our theoretical results of the resonant scattering by fractals. The exponent of the scattering enhancement found from our simulations equal $d_s=1.4\pm0.1$ for the random walk model and $d_s=1.3\pm0.1$ for the cluster-cluster aggregates. The value of the enhancement factor F_s obtained is of the order of 10^5 for the number of particles $N\sim(kR_0)^D\sim10^2$ and for the quality of resonance with a realistic value of $R_0^3|\mathrm{Im}\chi_0^{-1}|=0.001$. For example, clusters consisting of metal spherical particles

$$R_0^3|\mathrm{Im}\chi_0^{-1}|=\frac{R_0^3}{R_m^3}\frac{3\epsilon''}{|\epsilon-1|^2}\sim10^{-3}$$

for the visible and infrared regions of the spectrum.

Thus, resonant Rayleigh scattering of light by collective excitations of a fractal may be a good method of determining the optical spectral dimension, which gives information on dynamical properties of fractals. The study of both resonant and nonresonant scattering simultaneously allows one to obtain both the fractal dimension D, characterizing geometrical properties of fractals, and the optical spectral dimension d_0, which is an important characteristic of dynamics of fractals.

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*Also at the Université Paris-Sud, France, Physikalisches Institut der Universität Heidelberg, Germany, and L. V. Kirensky Institute of Physics, Krasnoyarsk, U.S.S.R.

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